FORM PTO-1390 (REV 10-95) | U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE | ATTORNEY'S DOCKET NUMBER: MERCK 2281

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. §371

U.S. APPLICATION NO. (If known, see 37 CFR §1.5): 09/889427

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/EP00/00069	7 JANUARY 2000	19 JANUARY 1999

TITLE OF INVENTION: MULTILAYER PEARL LUSTRE PIGMENT

OIPE JUL 18 2001 PATENT & TRADEMARK OFFICE

APPLICANT(S) FOR DO/EO/US: ANDES, Stephanie, et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ■ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. §371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. §371.
3. ☐ This express request to begin national examination procedures (35 U.S.C. §371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. §371(b) and PCT Articles 22 and 39(1).
4. ■ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ■ A copy of the International Application as filed (35 U.S.C. §371(c)(2))
   a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
   b. ■ has been transmitted by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ■ A translation of the International Application into English (35 U.S.C. §371(c)(2)).
7. ■ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. §371(c)(3))
   a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☐ have been transmitted by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ■ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. §371(c)(3)).
9. ■ An oath or declaration of the inventor(s) (35 U.S.C. §371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. §371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 C.F.R. §§1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 C.F.R. §§3.28 and 3.31 is included.
13. ■ A FIRST preliminary amendment.
    ☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☐ Other items or information:

U.S. APPLICATION NO. (if known, see 37 CFR 1.5)	INTERNATIONAL APPLICATION NO.	ATTORNEY'S DOCKET NUMBER
09/889427	PCT/EP00/00069	MERCK 2281

17. ☒ The following fees are submitted:	CALCULATIONS	PTO USE ONLY
BASIC NATIONAL FEE (37 CFR §1.492 (a) (1) - (5)):		
Search Report has been prepared by the EPO or JPO $860.00		
International preliminary examination fee paid to USPTO (37 CFR §1.482) $690.00		
No international preliminary examination fee paid to USPTO (37 CFR §1.482) but international search fee paid to USPTO (37 CFR §1.445(a)(2)) $710.00		
Neither international preliminary examination fee (37 CFR §1.482) nor international search fee (37 CFR §1.445(a)(2)) paid to USPTO $1000.00		
International preliminary examination fee paid to USPTO (37 CFR §1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) $100.00		
ENTER APPROPRIATE BASIC FEE AMOUNT =	$860.00	
Surcharge of **$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 C.F.R. §1.492(e)).		

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	12 - 20 =	0	x	$ 18.00	$0.00
Independent claims	2 - 3 =	0	x	$ 80.00	$0.00
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+	$ 270.00	
TOTAL OF ABOVE CALCULATIONS =					$860.00
Reduction of 1/2 for filing by small entity, if applicable. A Verified Small Entity Statement must also be filed (Note 37 C.F.R. §§1.9, 1.27, 1.28).					
SUBTOTAL =					$860.00
Processing fee of **$130.00** for furnishing the English translation later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 C.F.R. §1.492(f)).					
TOTAL NATIONAL FEE =					$860.00
Fee for recording the enclosed assignment (37 C.F.R. §1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 C.F.R. §§3.28, 3.31). $40.00 per property.					
TOTAL FEES ENCLOSED =					$860.00
				Amount to be refunded:	
				charged:	

a. ■ A check in the amount of $860.00 to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account No. 13-3402 in the amount of $ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ■ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 13-3402. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 C.F.R. §§1.494 or 1.495 has not been met, a petition to revive (37 C.F.R. §1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO: Customer Number 23,599

23599
PATENT TRADEMARK OFFICE

Filed: 18 JULY 2001

AJZ:kmo

SIGNATURE Reg # 32,004

Anthony J. Zelano
NAME

27,969
REGISTRATION NUMBER

APPLICATION DATA SHEET

APPLICATION INFORMATION

Application Type::	REGULAR
Subject Matter::	UTILITY
CD-ROM or CD-R?::	NONE
Title::	MULTILAYER PEARL LUSTRE PIGMENT
Attorney Docket Number::	MERCK 2281

INVENTOR INFORMATION

Inventor Authority Type::	INVENTOR
Primary Citizenship Country::	Germany
Status::	FULL CAPACITY
Given Name:: 2-00	Stephanie
Family Name::	ANDES
City of Residence::	Hanau DEX
Country of Residence::	Germany
Street::	Gustav-Hoch-Strasse 21
City::	Hanau
Country::	Germany
Postal or Zip Code::	D-63452

Inventor Authority Type::	INVENTOR
Primary Citizenship Country::	GERMANY
Status::	FULL CAPACITY
Given Name:: 3-00	Gerald
Family Name::	FUCHS-POHL
City of Residence::	Weiterstadt DEX
Country of Residence::	GERMANY
Street::	Arheilger Weg 2
City::	Weiterstadt
Country::	GERMANY
Postal or Zip Code::	D-64331

Inventor Authority Type::	INVENTOR
Primary Citizenship Country::	GERMANY
Status::	FULL CAPACITY
Given Name:: 1-00	Gerhard
Family Name::	PFAFF
City of Residence::	Munster DEX
Country of Residence::	GERMANY
Street::	Trautenauer Strasse 41
City::	Munster
Country::	GERMANY
Postal or Zip Code::	D-64839

CORRESPONDENCE INFORMATION

Correspondence Customer Number:: 23599

REPRESENTATIVE INFORMATION

Representative Customer Number:: 23599

FOREIGN PRIORITY INFORMATION

Country::	PCT
Application Number::	PCT/EP00/00069
Filing Date::	01/07/00
Country::	GERMANY
Application Number::	199 01 612.7
Filing Date::	01/19/99

ASSIGNMENT INFORMATION

Assignee Name::	MERCK PATENT GMBH
Street::	Frankfurter Strasse 250
City::	Darmstadt
Country::	Germany
Postal or Zip Code::	D-64293

IN THE UNITED STATES DESIGNATED/ELECTED OFFICE

International Application No.	:	PCT/EP00/00069
International Filing Date	:	7 JANUARY 2000
Priority Date(s) Claimed	:	19 JANUARY 1999
Applicant(s) (DO/EO/US)	:	ANDES, Stephanie, et al.

Title: MULTILAYER PEARL LUSTRE PIGMENT

PRELIMINARY AMENDMENT

Commissioner for Patents
Washington, D.C. 20231

SIR:

Prior to calculating the national fee, and prior to examination in the National Phase of the above-identified International application, please amend as follows:

IN THE CLAIMS:

4. (Amended) Pearl lustre pigment according to Claim 1, characterized in that the material of high refractive index is TiO_2, ZrO_2, Fe_2O_3, SnO_2, ZnO or a mixture of these oxides or an iron titanate, an iron oxide hydrate, a titanium suboxide or a mixture and/or mixed phase of these compounds.

7. (Amended) Process according to Claim 5, characterized in that the precursor is titanium tetrachloride.

8. (Amended) Process according to Claim 5, characterized in that following drying of the material to be coated the layers are applied in a fluidized-bed reactor by CVD and/or PVD.

9. (Amended) Use of the pigments according Claim 1 for pigmenting paints, printing inks, plastics cosmetics, glazes for ceramics, and glasses.

10. (Amended) Use of the pigments according to Claim 1 for the security sector, especially for printing items of value and of security, for agricultural films and for the laser marking of plastics.

11. (Amended) Paints, printing inks, plastics, cosmetics, ceramics, glasses and polymer films pigmented with a pigment according to Claim 1.

12. (Amended) Laser-markable plastics comprising pigments according to Claim 1.

09530247.072801

REMARKS

The purpose of this Preliminary Amendment is to eliminate multiple dependent claims in order to avoid the additional fee. Applicants reserve the right to reintroduce claims to canceled combined subject matter.

Respectfully submitted,

Reg. # 32,004

Anthony J. Zelano, Reg. No. 27,969
Attorney for Applicants
MILLEN, WHITE, ZELANO & BRANIGAN, P.C.
Arlington Courthouse Plaza 1
2200 Clarendon Boulevard, Suite 1400
Arlington, VA 22201
Direct Dial: 703-812-5311
Facsimile: 703-243-6410
Email: zelano@mwzb.com

AJZ:jmm

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VERSION WITH MARKINGS TO SHOW CHANGES MADE

Claims 4 and 7-12 have been amended as follows:

4. (Amended) Pearl lustre pigment according to ~~at least one of~~ Claims ~~1 to 3~~, characterized in that the material of high refractive index is TiO_2, ZrO_2, Fe_2O_3, SnO_2, ZnO or a mixture of these oxides or an iron titanate, an iron oxide hydrate, a titanium suboxide or a mixture and/or mixed phase of these compounds.

7. (Amended) Process according to ~~at least one of~~ Claims ~~5 and 6~~, characterized in that the precursor is titanium tetrachloride.

8. (Amended) Process according to ~~at least one of~~ Claims ~~5 to 7~~, characterized in that following drying of the material to be coated the layers are applied in a fluidized-bed reactor by CVD and/or PVD.

9. (Amended) Use of the pigments according ~~to~~ Claims ~~1 to 4~~ for pigmenting paints, printing inks, plastics cosmetics, glazes for ceramics, and glasses.

10. (Amended) Use of the pigments according to Claims ~~1 to 4~~ for the security sector, especially for printing items of value and of security, for agricultural films and for the laser marking of plastics.

11. (Amended) Paints, printing inks, plastics, cosmetics, ceramics, glasses and polymer films pigmented with a pigment according to Claims ~~1 to 4~~.

12. (Amended) Laser-markable plastics comprising pigments according to Claims ~~1 to 4~~.

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JC17 Rec'd PCT/PTO 18 JUL 2001

Multilayer pearl lustre pigment

The invention relates to a multilayer pearl lustre pigment having a pronounced colour flop, based on a platelet-shaped substrate comprising a material having a refractive index of more than 1.8.

Multilayer pigments which exhibit an angle-dependent colour change between two or more intensive interference colours are known.

For instance, US 4,434,010 describes a multilayer interference pigment consisting of a central layer of a reflective material (aluminium) and alternating layers of two transparent, dielectric materials of high and low refractive index, for example titanium dioxide and silicon dioxide, on either side of the central aluminium layer. In a further embodiment of the pigment, the layers following the central aluminium layer are formed by magnesium fluoride and chromium. This pigment exhibits an intensive colour flop from green to purplish red.

EP 0 753 545 describes goniochromatic lustre pigments based on transparent, non-metallic, platelet-shaped substrates, which have at least one layer stack comprising a colourless coating with a refractive index $n \leq 1.8$ and a reflective, selectively or non-selectively absorbing coating which is at least partly transparent to visible light, and which also have, if desired, an external protective layer in addition.

These pigments have the disadvantage that they are produced by a technically very complex and costly process, for example by chemical vapour deposition (CVD) or physical vapour deposition (PVD) techniques. Further disadvantages are the frequent difficulty in reproducing the pigments in the desired product quality, and their deficient weathering stability.

It is the object of the present invention to provide an essentially transparent interference pigment having strong interference colours and/or a high angular dependency of the interference colours and featuring advantageous applications properties, which at the same time is simple to produce.

This object is achieved in accordance with the invention by a multilayer pearl lustre pigment on the basis of a platelet-shaped substrate comprising a material having a refractive index of more than 1.8, which comprises at least

(i) a first layer of a material of low refractive index in the range from 1.35 to 1.8,

(ii) optionally, a second layer of a material having a refractive index of more than 1.8,

(iii) a semitransparent metal layer which is applied to the substrate or to the layers (i) or (ii), and

(iv) if desired, an aftercoating.

If the semitransparent metal layer forms the outer layer of the pigment, it is also possible for layers of high and low refractive index to follow. Before the metal layer is applied, the first and second layers may also be repeated.

This object is further achieved, in accordance with the invention, by a process for producing the pigment of the invention by

- applying a precursor of the substrate material as a thin film to a continuous belt,

- solidifying the liquid film by drying and, in so doing, developing the metal oxide by chemical reaction from the precursor,
- detaching the dried film,
- washing the resultant substrate particles and resuspending them in a coating solution,
- coating the substrate particles with two or more layers of metal oxides or metals, and
- aftercoating the resultant pigment.

Alternatively, the layer system can be produced with the aid of a PVD technique or by a combination of wet-chemical techniques and/or CVD and/or PVD techniques.

The invention additionally provides for the use of the pigments of the invention in paints, varnishes, printing inks, plastics, ceramic materials, glasses and cosmetic formulations. For these purposes they may also be employed as mixtures with commercially customary pigments, examples being organic and inorganic absorption pigments, metal-effect pigments and LCP pigments.

In addition to the purely colouristic applications, the pigments of the invention can also be considered for functional applications. Examples of these are as pigments for the security sector, e.g. the printing of items of value and of security, as pigments with specific IR reflection, e.g. for glasshouse films, and as pigments for the laser marking of plastics.

The pigments of the invention are based on platelet-shaped substrates having a refractive index of more than 1.8. These substrates may consist, for example, of titanium dioxide, zirconium dioxide, α-iron(III) oxide, tin oxide, zinc oxide or other transparent and stable materials capable of taking on soluble or insoluble colorants.

Precursors employed for the production of the substrates are solutions of organic or inorganic compounds of the metals titanium, zirconium, iron, tin, zinc or mixtures thereof. A preferred precursor is titanium tetrachloride.

The platelet-shaped substrate particles have a thickness of between 0.05 and 5 µm and, in particular, between 0.05 and 2 µm. The extent in the other two dimensions is between 2 and 200 µm, and, in particular, between 5 and 50 µm.

Suitable layer material for the layer (i) having a refractive index of from 1.35 to 1.8 comprises all materials of low refractive index which are known to the person skilled in the art and can be applied permanently and in film-like manner to the substrate particles. Particularly suitable are metal oxides or metal oxide mixtures, such as SiO_2, Al_2O_3, AlO(OH), B_2O_3 or a mixture of the said metal oxides or MgF_2. Alternatively, the material of low refractive index employed can comprise polymers, such as acrylates. The monomers used have a molecular weight of from 200 to 1000 and are available as mono-, di- or triacrylates. In terms of functional groups, they are available as hydrocarbons, polyols, polyethers, silicones or fluorinated Teflon-like monomers. These monomers can be polymerized by electron beams or UV rays. The layers obtained possess a temperature stability of up to 250°C. The refractive indices of the acrylate layers lie within the range from 1.35 to 1.60. Further details can be found in David G. Shaw and Marc G. Langlois: Use of a new high speed acrylate deposition process to make novel multilayer structures, MRS Conference in San Francisco 1995; A new high speed process for vapour depositing fluoro and silicone acrylates for release coating applications, Conference of the Society of Vacuum Coaters in Chicago, Illinois, 1995.

The thickness of the layer (i) is 10 - 1000 nm, preferably 20 - 800 nm and, in particular, 30 - 600 nm.

Suitable layer materials for the layer (ii) having a refractive index of more than 1.8 are preferably metal oxides or metal oxide mixtures, such as TiO_2, Fe_2O_3, ZrO_2, ZnO, SnO_2, or compounds of high refractive index such as iron titanates, iron oxide hydrates, titanium suboxides, chromium oxide, bismuth vanadate, cobalt aluminate, and also mixtures and/or mixed phases of the said compounds with one another or with other metal oxides. Metal sulphides, metal nitrides and metal oxynitrides are also suitable. The thickness of the layer (ii) is 10 - 550 nm, preferably 15 - 400 nm and, in particular, 20 - 350 nm.

The metal layers (iii) consist of metals, such as aluminium, chromium, nickel, chromium-nickel alloys or silver. Chromium and aluminium are preferred here, since they are easy to deposit. The layer thickness of the metal layers is set at from 5 to 20 nm in order to obtain semitransparency. Alternatively, materials such as graphite or titanium nitride can be employed as semitransparent reflector layers.

The pigments of the invention also include additional colorants in the coating. If, for example, particles of carbon black are used, then particle sizes of from 5 to 200 nm, and, in particular, from 10 to 100 nm are used. Pigments of this kind, which contain preferably carbon black particles in layers of titanium dioxide, iron oxide, tin oxide, chromium oxide and zinc oxide, are described in EP 0 499 864.

In addition, the pigments of the invention may also comprise particles of titanium dioxide, aluminium oxide, silicon dioxide, tin dioxide, magnesium oxide, zinc oxide, cerium dioxide, tungsten oxide, molybdenum

oxide, zirconium oxide, or else mixed oxides, such as Cr_2FeO_4, $CoAl_2O_4$ or $NiAl_2O_4$, in the coating.

Instead of inorganic pigment particles it is also possible for organic pigment particles to be present in the coating, in which case particular preference is given to temperature-stable organic pigments. Organic pigment particles used are preferably phthalocyanines, products of laking basic dyes with heteropolyacids, and anthraquinones, phenazines, phenoxazines, diketopyrrolopyrroles or perylenes. In principle, all pigments which have been described for incorporation into the substrate can also be incorporated into the coating of the pigment of the invention. The incorporation of small particles of metal oxide or organic pigment having an average size of from 10 to 40 nm into the cavities of the metal oxide coating brings about a marked increase in the hiding power and in the lustre, in association with a high level of homogeneity of the coating in comparison to pigments obtained by coprecipitation. The hiding power and, in the case of coloured pigment particles, the observation-angle-dependent absorption colour of the pigments of the invention can be varied within a wide range by way of the concentration of the pigment particles incorporated. The mass fraction of incorporated pigment particles, based on the coating, lies between 0.5 and 30% and, in particular, between 2 and 20%. Further details of pigments which comprise pigment particles in the coating can be found in DE 41 40 295.

The finished pigment can be subjected to an aftercoating or aftertreatment (iv), which increases further the light stability, weathering stability and chemical stability, or which facilitates the handling of the pigment, especially its incorporation into various media. Suitable aftercoatings or aftertreatments are, for example, the processes

described in DE-C 22 15 191, DE-A 31 51 354, DE-A 32 35 017 or DE-A 33 34 598.

The additionally applied substances account for only from about 0.1 to 5% by weight, preferably from about 0.5 to 3% by weight, of the overall pigment.

The number and thickness of the layers is dependent on the desired effect and on the substrate used. The number of layers is limited by the economics of the pigment. If the substrate used is TiO_2 platelets, which in accordance with the process described in WO 97/43346 are produced on a continuous belt, it is possible to obtain particularly well-defined interference effects, since these TiO_2 platelets possess a uniform layer thickness. The reflection spectrum or transmission spectrum of such a pigment features finer and more precisely harmonizable structures than the spectrum of a corresponding pigment which is based on a substrate having a broad thickness distribution, such as mica, for example.

In accordance with WO 97/43346 the TiO_2 platelets are produced on a continuous belt by solidification and hydrolysis of a titanium tetrachloride solution.

The metal oxide layers are preferably applied by wet-chemical means, it being possible to employ the wet-chemical coating techniques developed for the production of pearl lustre pigments; such techniques are described, for example, in DE 14 67 468, DE 19 59 988, DE 20 09 566, DE 22 14 545, DE 22 15 191, DE 22 44 298, DE 23 13 331, DE 25 22 572, DE 31 37 808, DE 31 37 809, DE 31 51 343, DE 31 51 354, DE 31 51 355, DE 32 11 602, DE 32 35 017 or else in further patent documents and in other publications.

For coating, the substrate particles are suspended in water and the suspension is admixed with one or more

hydrolysable metal salts at a pH suitable for the hydrolysis, this pH being chosen such that the metal oxides and/or metal oxide hydrates are deposited directly on the particles without instances of secondary precipitation. The pH is normally held constant by simultaneous metered addition of a base. Subsequently, the pigments are separated off, washed and dried and, if desired, calcined, it being possible to optimize the calcination temperature in respect of the particular coating present. If desired, the pigments can be separated off, dried and, if desired, calcined following the application of individual coatings, before then being resuspended in order to apply the further layers by precipitation.

In addition, coating can also be carried out by gas-phase coating in a fluidized-bed reactor, it being possible to employ, accordingly, the techniques proposed in EP 0 045 851 and EP 0 106 235 for the production of pearl lustre pigments.

For the application of titanium dioxide layers, preference is given to the technique described in US 3,553,001.

An aqueous titanium salt solution is added slowly to a suspension, heated to about 50-100°C, especially 70-80°C, of the material to be coated, and a substantially constant pH of about 0.5-5, in particular about 1.5-2.5, is maintained by simultaneous metered addition of a base, such as aqueous ammonia solution or aqueous alkali metal hydroxide solution, for example. As soon as the desired layer thickness of the TiO_2 precipitate is reached, the addition of the titanium salt solution and of the base is stopped.

This technique, which is also referred to as the titration technique, is notable for the fact that it avoids an excess of titanium salt. This is achieved by

supplying to the hydrolysis per unit time only that quantity of titanium salt solution which is required for uniform coating with the hydrated TiO_2 and can be received per unit time by the available surface area. Consequently, no hydrated titanium dioxide particles are produced that are not precipitated on the surface to be coated.

For the application of the silicon dioxide layers, the following technique can be employed: a sodium waterglass solution is metered into a suspension, heated at about 50-100°C, especially 70-80°C, of the material to be coated. The pH is held constant at from 4 to 10, preferably from 6.5 to 8.5, by simultaneous addition of 10% hydrochloric acid. Following the addition of the waterglass solution, stirring is continued for 30 minutes.

The individual layers can also be produced in accordance with known techniques by sputtering metals, such as aluminium or chromium, or alloys, such as Cr-Ni alloys, and also metal oxides, for example titanium oxide, silicon oxide, or indium-tin oxide, or by thermal evaporation of metals, metal oxides or acrylates. Preference is given to a vacuum belt coating as described in DE 197 07 805 and in DE 197 07 806 for the production of interference pigments.

Patent Claims

1. Multilayer pearl lustre pigment on the basis of a platelet-shaped substrate comprising a material having a refractive index of more than 1.8, which comprises at least

   (i) a first layer of a material of low refractive index in the range from 1.35 to 1.8,

   (ii) optionally, a second layer of a material having a refractive index of more than 1.8,

   (iii) a semitransparent metal layer which is applied to the substrate or to the layers (i) or (ii), and

   (iv) if desired, an aftercoating.

2. Pearl lustre pigment according to Claim 1, characterized in that the substrate is platelet-shaped titanium dioxide, zirconium dioxide, α-iron(III) oxide, tin dioxide or zinc oxide.

3. Pearl lustre pigment according to Claims 1 and 2, characterized in that the material of low refractive index is SiO_2, Al_2O_3, AlO(OH), B_2O_3, MgF_2 or an acrylate.

4. Pearl lustre pigment according to at least one of Claims 1 to 3, characterized in that the material of high refractive index is TiO_2, ZrO_2, Fe_2O_3, SnO_2, ZnO or a mixture of these oxides or an iron titanate, an iron oxide hydrate, a titanium suboxide or a mixture and/or mixed phase of these compounds.

5. Process for producing the pigment of the invention by

- applying a precursor of the substrate material as a thin film to a continuous belt,
- solidifying the liquid film by drying and, in so doing, developing the metal oxide by chemical reaction from the precursor,
- detaching the dried film,
- washing the resultant substrate particles and resuspending them in a coating solution,
- coating the substrate particles with two or more layers of metal oxides or metals, and
- aftercoating the resultant pigment.

6. Process according to Claim 5, characterized in that precursors employed are solutions of organic or inorganic compounds of the metals titanium, zirconium, iron, tin or zinc.

7. Process according to at least one of Claims 5 and 6, characterized in that the precursor is titanium tetrachloride.

8. Process according to at least one of Claims 5 to 7, characterized in that following drying of the material to be coated the layers are applied in a fluidized-bed reactor by CVD and/or PVD.

9. Use of the pigments according to Claims 1 to 4 for pigmenting paints, printing inks, plastics, cosmetics, glazes for ceramics, and glasses.

10. Use of the pigments according to Claims 1 to 4 for the security sector, especially for printing items of value and of security, for agricultural films and for the laser marking of plastics.

DE19707215990

11. Paints, printing inks, plastics, cosmetics, ceramics, glasses and polymer films pigmented with a pigment according to Claims 1 to 4.

12. Laser-markable plastics comprising pigments according to Claims 1 to 4.

Abstract

Multilayer pearl lustre pigment on the basis of a platelet-shaped substrate comprising a material having a refractive index of more than 1.8, which comprises at least

(i) a first layer of a material of low refractive index in the range from 1.35 to 1.8,

(ii) optionally, a second layer of a material having a refractive index of more than 1.8.

(iii) a semitransparent metal layer which is applied to the substrate or to the layers (i) or (ii), and

(iv) if desired, an aftercoating, the substrate being platelet-shaped titanium dioxide, zirconium dioxide, α-iron(III) oxide, tin dioxide or zinc oxide.

Docket No.
Merck

Declaration and Power of Attorney For Patent Application
English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

MULTILAYER PEARL LUSTRE PIGMENT

the specification of which

(check one)

☐ is attached hereto.
☐ was filed on 07.01.2000 as United States Application No. or PCT International Application Number PCT/EjP00/00069
and was amended on _____
(if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119(a)-(d) or Section 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)			Priority Not Claimed
199 01 612.7	DE	19.01.1999	☐
(Number)	(Country)	(Day/Month/Year Filed)	
			☐
(Number)	(Country)	(Day/Month/Year Filed)	
			☐
(Number)	(Country)	(Day/Month/Year Filed)	

I hereby claim the benefit under 35 U.S.C. Section 119(e) of any United States provisional application(s) listed below:

(Application Serial No.)	(Filing Date)
(Application Serial No.)	(Filing Date)
(Application Serial No.)	(Filing Date)

I hereby claim the benefit under 35 U.S.C. Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States of PCT International application in the manner provided by the first paragraph of 35 U.S.C. Section 112. I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, C.F.R., Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application:

(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)
(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)
(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

POWER OF ATTORNEY: As a named inventor, I hereby appoint

I. William Millen (Reg. No. 19,544)
John L. White (Reg. No. 17,746)
Anthony J. Zelano (Reg. No. 27,969)
Alan E.J. Branigan (Reg. No. 20,565)
John R. Moses (Reg. No. 24,983)
Harry B. Shubin (Reg. No. 32,004)
Brion P. Heaney (Reg. No. 32,542)
Richard J. Traverso (Reg. No. 30,595)
John A. Sopp (Reg. No. 33,103)
Richard M. Lebovitz (Reg. No. 37,067)
John H. Thomas (Reg. No. 33,460)
Catherine M. Joyce (Reg. No. 40,668)
James T. Moore (Reg. No. 35,619)
James E. Ruland (Reg. No. 37,432)
Nancy Axelrod (Reg. No. 44,014)
Jennifer J. Branigan (Reg. No. 40,921)

(16)

to prosecute this application and transact all business in the Patent and Trademark Office connected therewith.

Send Correspondence to: MILLEN; WHITE; ZELANO & BRANIGAN, P.C.
Arlington Courthouse Plaza I
2200 Clarendon Blvd., Suite 1400
Arlington, VA 22201

Direct Telephone Calls to: (name and telephone number)

1-00

Full name of sole or first inventor	
PFAFF, Gerhrad	21.05.2001
Sole or first inventor's signature	Date
Residence	
Citizenship	
Post Office Address	
c/o Merck KGaA, Frankfurter Str. 250, 64293 Darmstadt, Germany	

2-00

Full name of second inventor, if any	
ANDES, Stephanie	21.05.2001
Second inventor's signature	Date
Residence	
Citizenship	
Post Office Address	
c/o Merck KGaA, Frankfurter Str. 250, 64293 Darmstadt, Germany	

3-00

Full name of third inventor, if any	
FUCHS-POHL, Gerald	21.05.2001
Third inventor's signature	Date
Residence	
Citizenship	
Post Office Address	
c/o Merck KGaA, Frankfurter Str. 250, 64293 Darmstadt, Germany	

Full name of fourth inventor, if any	
Fourth inventor's signature	Date
Residence	
Citizenship	
Post Office Address	

Full name of fifth inventor, if any	
Fifth inventor's signature	Date
Residence	
Citizenship	
Post Office Address	

Full name of sixth inventor, if any	
Sixth inventor's signature	Date
Residence	
Citizenship	
Post Office Address	